

PCT

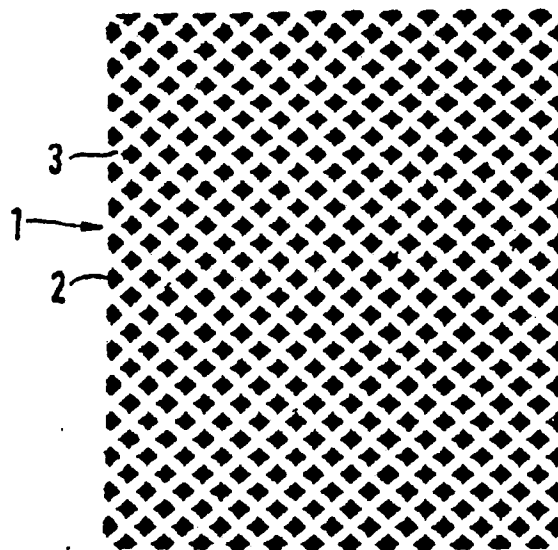
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<p>(21) International Application Number: PCT/US89/05804 (22) International Filing Date: 22 December 1989 (22.12.89) (30) Priority data: P 38 44 250.7 29 December 1988 (29.12.88) DE (71) Applicant (for all designated States except US): MINNESOTA MINING AND MANUFACTURING COMPANY [US/US]; P.O. Box 33427, Saint Paul, MN 55133 (US). (72) Inventors; and (75) Inventors/Applicants (for US only) : JUNG, Dieter [DE/DE]; VON JAKUSCH, Egbert [DE/DE]; KRECKEL, Karl-Werner [DE/DE]; Wilbecke 12-14, P.O. Box 1340, D-4280 Borken 1 (DE). (74) Agents: SPRAGUE, Robert, W. et al.; Minnesota Mining and Manufacturing Company, P.O. Box 33427, St. Paul, MN 55133 (US).</p>		<p>(81) Designated States: AT (European patent), AU, BE (European patent), CH (European patent), DE (European patent), ES (European patent), FR (European patent), GB (European patent), IT (European patent), JP, KP, LU (European patent), NL (European patent), SE (European patent), US.  Published With international search report.</p>

(54) Title: PATTERNED SILICONE RELEASE COATED ARTICLE



(57) Abstract

A primerless release article comprising a film substrate, having coated on at least a portion thereof, at least one layer of a printable silicone release agent, wherein the agent forms a definite geometric pattern consisting of discrete islands and bridges, the islands having dimensions of from about 0.25 mm by 0.25 mm to about 4 mm by 4 mm, the bridges forming a grid, each of the bridges having a width of from about 0.25 mm to about 0.75 mm.

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## PATTERNED SILICONE RELEASE COATED ARTICLE

Field of the Invention

5           The invention relates to a substrate which has coated to at least a portion thereof, at least one release agent, wherein the release agent forms a definite geometric pattern.

Background of the Invention

10           Silicone release agents are well known in the art. Numerous silicones are commercially available, as are liners coated with a layer of silicone. However, these silicones have various drawbacks including controlling the  
15 amount of release desirable. Frequently, silicones provides what is known in the art as "premium release", meaning that they exhibit extremely low release values, i.e., 4-60 g/cm of width. These low values are not always desirable, especially for release coatings to be used on  
20 the back surfaces of adhesive tapes. This type of back surface coating is known as "low adhesion backsize" or LAB. A controlled release having exhibiting values of from 60 to about 500 g/cm of width to the adhesive is desired, so that the tape will not prematurely unwind. Silicones are  
25 available in both solvent curable, and uv-curable systems. Many nonsilicone polymers such as urethanes are used as LABs because of their higher release force than the silicones. Various modifications of silicones have been attempted for use in adhesive tapes, such as binding them  
30 with polymers having higher releases, as in U.S. Pat. Nos. 3,3288,482, 3,527,659 and the like.

          Pattern release coatings have been of interest for many years for the goal of controlling release in silicones. In DE-PS 8 55 746, the idea of making a release  
35 coating which formed a pattern was disclosed. However, no method for accomplishing the pattern coating was disclosed,

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nor exemplified. Likewise, UK Patent Application GB 2,209,148 A, published May 4, 1989, discloses a two-layer self-adhesive label having a self-adhesive base which provides a non-adherent area. The nonadherent area is described as printed with a pattern UV cured release panel. However, no method for making such a pattern is disclosed or claimed.

In U.S. Pat. No. 3,503,782, a differential release paper is made by applying a full coating of a first release agent, and a second partial coating of a differing release agent. It is stated that the second coating may be provided in the form of a pattern. In this manner, the surface is provided with a variable release force. However, this requires the complete coating of the surface with one release agent, typically requiring several processing steps and waste of a certain amount of silicone release agent which is under area covered by the second agent.

In DE 3727 078 A1, Beiersdorf, published February 23, 1989, it is disclosed that the problems of making and using this type of coating have never been resolved. It is disclosed in this publication that two layer screen-printed silicone release coatings may be made on a substrate by priming the substrate with a commercial primer, known to have a low release value. Primers disclosed include isocyanatelinked polyvinyl acetate, and solutions of chromostearate complexes mixed with polyvinyl alcohol. The silicone may then be overcoated onto the primer. The silicone is coated using a screen with a sieve size of 10-450 mesh, and typically covers 20-80% of the surface of the primer.

Thus, all of the patterned release liners for which methods of manufacture are disclosed above, have required the use of a primer, or second fully coated layer of release agent.

Processes are also known for pattern printing of other compounds onto substrates, i.e., adhesives. In U.S. Pat. No. 3,741,786, pressure-sensitive adhesive transfer tapes are disclosed having a plurality of substantially

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non-contiguous raised pressure-sensitive adhesive segments. This construction is disclosed to allow the pressure-sensitive segments to be transferred.

An objective of the present invention is to  
5 provide a release article having a patterned coating for use with adhesive tapes and other applications which does not require the use of a primer with the silicone release agent.

Another objective of the present invention is to  
10 provide a plurality of articles having smooth controlled release varying from about 100 g/cm to well over 500 g/cm.

#### Summary of the Invention

The invention provides a primerless release  
15 article comprising a film substrate, having coated on at least a portion thereof, at least one layer of a printable silicone release agent, wherein said agent forms a definite geometric pattern consisting of discrete islands and  
20 bridges, said islands having dimensions of from about 0.25 mm by 0.25 mm to about 4 mm by 4 mm, said bridges forming a grid, each bridge having a width of from about 0.25 mm to about 1.0 mm.

More specifically, the invention provides two  
25 different types of primerless release articles, one being the negative of the other.

The invention provides one type of primerless  
release article comprising a film substrate, and coated on at least a portion thereof, at least one layer of a  
30 printable silicone release agent, wherein said agent forms a definite geometric pattern consisting of discrete coated islands and uncoated bridges, said coated islands having  
dimensions of from about 0.25 mm by 0.25 mm to about 4 mm by 4 mm, said uncoated bridges forming a grid, each bridge  
having a width of from about 0.25 mm to about 1.00 mm.

35 The invention also provides a second type of primerless release article comprising a film substrate, and coated on at least a portion thereof, at least one layer of

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a printable silicone release agent, wherein said agent forms a definite geometric pattern consisting of discrete uncoated islands and coated bridges, said uncoated islands having dimensions of from about 0.25 mm by 0.25 mm to about 4 mm by 4 mm, said coated bridges forming a grid, each bridge having a width of from about 0.25 mm to about 1.0 mm.

As used herein, the ensuing terms have the correlative meanings.

1. The term "island" means a discrete area having a defined geometric shape.

2. The term "bridge" means a discrete columnar area which may intersect with other bridges to form a grid pattern.

3. The term "grid" means a pattern of interconnected bridges emanating from differing points, forming and surrounding roughly equivalent geometric islands.

#### Description of the Drawings

FIG. 1 shows a release article of the invention. The surface of the substrate 1, is coated with a silicone release agent in a pattern characterized by coated areas, or islands 2, each island having an edge area of 3 mm and uncoated bridges 3 forming a grid, each bridge having a width of 1 mm. The total percentage of the surface area which is siliconized is 56%.

FIG. 2 shows another release article of the invention. The surface of the substrate 1, is coated with a silicone release agent in a pattern characterized by coated areas 2 and uncoated bridges 3 forming a grid. The islands are similar in size to those of Fig. 1, the bridges have a width of 0.5 mm, narrower than those of Figure 1. The total percentage of the surface area which is siliconized is 72%.

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FIG. 3 shows another release article of the invention. The surface of the substrate 1, is coated with a silicone release agent in a pattern characterized by coated areas 2 and uncoated bridges 3, forming a grid. This pattern is characterized by smaller islands than Figures 1 and 2, each island having an edge area of 2 mm, and each bridge width being 0.5 mm. The total percentage of the surface area which is siliconized is 64%.

FIG. 4 shows a second type of release article of the invention. The surface of the substrate 1, is coated by a silicone release agent in a pattern characterized by uncoated islands 3, each uncoated island having an edge area of 1 mm and coated bridges forming a grid 2, each bridge having a width of 0.75 mm. The total percentage of the surface area which is siliconized is 33%.

FIG. 5 shows another release article of the invention. The surface of the substrate 1, is coated with a silicone release agent in a pattern characterized by coated islands 2 and uncoated bridges 3, forming a grid. This pattern is characterized by smaller islands than Figures 1 and 2, each island having a edge area of 2 mm, and each bridge width being 0.75 mm. The total percentage of the surface area which is siliconized is 53%.

FIG. 6 shows another release article of the invention. The surface of the substrate 1, is coated with a silicone release agent in a pattern characterized by coated islands 2 and uncoated bridges 3, forming a grid. This pattern is characterized by square islands having an edge area of 1.75 mm, and each bridge width being 0.75 mm. The total percentage of the surface area which is siliconized is 49%.

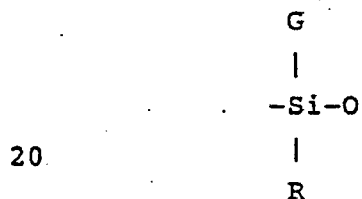
#### Detailed Description of the Invention

Release articles of the invention are particularly useful when release force values in excess of 100 g/cm are desired.

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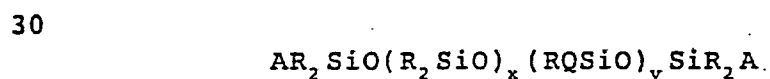
Silicone release agents useful in articles of the invention include a wide variety of organopolysiloxanes. The compositions are preferably, but not necessarily, high molecular weight silicone polymers or copolymers. Some  
 5 useful examples include both ultraviolet radiation cured and solvent cured silicones such as polyorganosiloxanes, including polydimethylsiloxanes, epoxypolysiloxanes, and the like.

In preferred articles, a solvent-free crosslink-  
 10 able silicone system is used, such the epoxy functional diorganopolysiloxanes, in which up to 20% of the groups are epoxy functional groups as described in U.S. Pat. 4,547,431. Also useful are the radiation curable coatings described in U.S. Patent No. 4,576,999 wherein there is  
 15 disclosed precrosslinking polyorganosiloxane containing units of



where R is C<sub>(1-3)</sub> alkyl and G is independently C<sub>(1-3)</sub> alkyl, an epoxy-functional organic radical of 2 to 20  
 25 carbon atoms, or an acrylic functional radical of 2-20 carbon atoms with at least one unit being epoxy or acrylic functional.

Also useful are epoxy substituted polysiloxanes having the formula

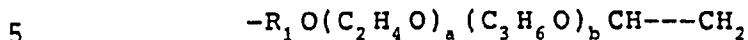


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wherein  $R_2$  is a monovalent hydrocarbon or halogenated hydrocarbon group having from 1 to 10 carbon atoms, A is R or Q, and Q is a group with the formula



O

$R_1$  is an alkylene group having from 3 to 5 carbon atoms, a has an average value of 0 to 300 and b has an average value of 0 to 30, a + b has an average value of 2 to 60, x has an average value of 1 to 500, and y has an average value of 0 to 100, and each molecule contains at least one Q group.

Also useful are combinations of vinyl endblocked diorganopolysiloxanes in which from 3 to 30 mole percent of the nonterminal siloxane units and at least 50 mole percent of the remaining organic radicals on the siloxane units are methyl radicals, and organopolysiloxanes which have at least 3 Si-bonded hydrogen atoms per molecule as disclosed in U.S. Pat. 4,208,504.

The release agent exhibits a definite geometric pattern consisting of discrete islands and bridges on the substrate, wherein the islands have dimensions of from about 0.25 mm by 0.25 mm to about 4 mm by 4 mm, and the uncoated bridges form a grid, each bridge having a width of from about 0.25 mm to about 1.0 mm. In preferred articles of the invention, the islands have dimensions of from about 0.50 mm by 0.50 mm to about 3 mm by 3 mm. Where a high tack adhesive is used, the bridges must be no wider than 0.75 mm, preferably from about 0.25 mm to about 0.50 mm.

The islands present in the pattern, whether coated areas, or uncoated areas, may be square, circular, triangular, elliptic, rhombic, or other defined geometric shape. The bridges then form a related grid. For example, where the islands are circular, the bridges are narrow between adjacent circles, and the intersection of the bridges is enlarged; where the islands are square, the bridges form diagonally opposed equal lines.

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The particular geometric shape used depends on the release values desired for the release article, the nature of the adhesive which it will contact, and the type of substrate.

5           Useful substrates are films, preferred substrates are thermosplastic films such as polyolefins, foils or other suitable nonporous tape backings.

10           The printing of the release coat or coats may be carried out in a single printing step by any suitable printing technique. The pattern is coated on a roll-coater having a plurality of transfer rolls equipped with feed devices which meter the release agent in accurately defined weights onto the outer roll surface of the coater roll. The coater rolls are fitted with an embossed outer  
15           surface corresponding to the pattern to be printed onto the substrate. The solvent-free systems, e.g. uv-curable materials are especially print-technology applicable.

20           Articles of the invention are useful for rolled adhesive tape, for abrasive articles, and for bandages for transcutaneous administration of a microemulsion medicament. Such a device contains an absorbant material placed in a stamped out reservoir which intersects a piece of closed-cell polyethylene foam. The medicament is contained in the absorbant material. The polyethylene foam  
25           piece is covered by a protective polyester foil for storage. This foil is coated with the patterned silicone coating of the invention.

30           The following tests have been used for evaluating articles of the invention. All weight, ratios, and percentages herein are by weight unless otherwise specifically noted.

#### Glossary of Terms

35           1. Tape X - A standard pressure-sensitive adhesive tape having an acrylic pressure-sensitive adhesive as disclosed in U.S. Patent No. Re 24,906, i.e.,

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a 95.5:4.5 isooctyl acrylate: acrylic acid adhesive coated onto a 0.5 mm polyester film.

2. Tape Y - A solvent based rubber resin pressure sensitive adhesive tape having 35% Kraton<sup>TM</sup> 1107, available from Shell Chemical Company, 16.5% Wingtack<sup>TM</sup> 10, available from Goodyear Chemical Company; and 46.5% Wingtack<sup>TM</sup> Plus, available from Goodyear Chemical Company, dispersed in a solvent mixture of 4:1 toluene:heptane, and coated onto a 90 mm cast polypropylene film.

3. Tape Z - A solvent based rubber resin pressure sensitive adhesive having 33% Kraton<sup>TM</sup> 1107; 44% Escorez<sup>TM</sup> 1310, available from Exxon Chemical Company; 22% Zonarez<sup>TM</sup> A25, available from Arizona Chemical and 1% Irganox<sup>TM</sup> 1076 Antioxidant, available from Ciba Geigy, dispersed in a mixture of 4:1 toluene:heptane and coated onto a polyethylene coated paper.

#### Test Methods

##### Release Value Test Procedure

A pressure-sensitive adhesive tape is pressed against the surface of a release coated substrate using two passes of a 2 Kg rubber roller to produce a laminate. The laminate was cut into 25 mm X 250 mm strips and attached, substrate side down, to a smooth stainless steel panel. The release value is the force, in grams, required to pull the pressure-sensitive adhesive tape with adhesive adhered thereto away from the release coated substrate at an angle of 180° and a pulling speed of 300 mm/min.

##### Heat Aged Release Value Test

A laminate of a pressure-sensitive adhesive tape and release coated substrate was prepared as described above and heated in an oven at 70°C under a constant load of 20 grams per square centimeter for 20 hours. After this time, the laminate was removed from the oven, allowed to cool for at least 10 minutes at room temperature, and within 2 hours after removal from the oven, the force

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required to pull the pressure sensitive tape with adhesive adhered thereto away from the release coated substrate was measured as described above in the test for Release Value.

## Readhesion Test

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A 25 X 250 mm strip of pressure sensitive tape, as removed from the release coated substrate, was pressed to the surface of a bright annealed stainless steel panel using two passes of a 2 Kg rubber roller. The readhesion value is the force, in Newtons/25 mm, required to pull the tape from the panel surface at an angle of 180° and a stripping speed of 300 mm/min.

### Examples

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### Example 1

An epoxy silicone having an epoxy equivalent weight of 865 is prepared as described in U.S. Patent No. 4,822,687, and coated onto an 0.085 mm cast polypropylene film in an island having 1 mm x 1 mm islands and separated by bridges of 0.375 mm. The silicone is cured as described in the reference. The sample was tested according to the test methods for Release Value, Heat-Aged Release Value, and Readhesion, described above and the test results are summarized in Table 1.

### Examples 2-10

Examples 2-10 were prepared as described in Example 1 except that the island and bridge sizes were changed as indicated in Table 1.

Examples 12-14

Examples 12-14 were made as described in Example 1 with island and bridge sizes as shown in Table 1. Test  
35 Tape Y was used in the Release Value Test.

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Examples 15-17

Examples 15-17 were made as using an epoxy silicone having an Epoxy Equivalent Weight of 445. The epoxy silicone was made as described in U.S. Pat.No. 4,822,687. The island size and bridge widths are shown in Table 1.

Examples 18-30

Examples 18-30 were made by coating Wacker-Chemie "VP-1530", available from Wacker-Chemie GmbH in the island sizes and bridge width as indicated in Table 1 and cured under UV light.

Examples 31-32

Examples 31-32 were made by coating Wacker Dehasive™ 920, available from Wacker-Chemie GmbH in the island sizes and bridge width as indicated in Table 1 and heat cured.

Examples 33-35

Examples 33-35 were made as in Example 1 except that the island was coated as a grid with the island sizes indicating the uncoated islands and the bridge sizes indicating the width of the coated bridges forming the grid.

Example 36

Example 36 was made as in Example 33 except that the silicone was coated onto a 0.040 mm biaxially oriented polypropylene film.

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TABLE I

Ex	Islands Size-mm	bridge mm	Silicone/ Test	Substrate Tape	Release		Heat Aged		Readhe-	
					Value	g/25mm	Value	g/25mm	sion	Value
										N/25mm
1	1X1	0.375	A	X	242		234			13.4
2	1X1	0.500	A	X	312		319			13.1
3	1X1	0.625	A	X	429		452			11.4
4	2X2	0.375	A	X	137		174			11.8
5	2X2	0.500	A	X	139		176			13.4
6	2X2	0.625	A	X	385		450			9.9
7	3X3	0.375	A	X	54		51			14.9
8	3X3	0.500	A	X	79		95			11.9
9	3X3	0.625	A	X	230		261			9.8
10	4X4	0.500	A	X	80		62			16.5
11	Comparative*		A	X	10		15			14.5
12	1X1	0.025	A	Y	77		---			---
13	1X1	0.500	A	Y	791		---			---
14	1X1	0.750	A	Y	1507		---			---
15	1X1	0.025	B	Y	533		---			---
16	1X1	0.500	B	Y	1039		---			---
17	1X1	0.750	B	Y	1813		---			---
18	1X1	0.375	C	Z	49		349			12.1

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19	1X1	0.500	C	Z	126	648	11.5
20	1X1	0.625	C	Z	81	360	12.0
21	2X2	0.375	C	Z	17	138	15.3
22	2X2	0.500	C	Z	41	211	15.4
23	2X2	0.625	C	Z	64	392	11.5
24	3X3	0.375	C	Z	12	87	15.9
25	3X3	0.500	C	Z	18	109	14.9
26	3X3	0.625	C	Z	32	161	13.7
27	Comparative*		C	Z	15	18	17.5
28	2X2	0.750	C	#5336	495	804	---
29	1.75X1.75	0.750	C	#5336	532	968	---
30	Comparative*		C	#5336	72	91	---
31	2X2	0.750	D	#5337	1196	1826**	40.5
32	Comparative*		D	#5337	740	208**	47.2
33***	1X1	0.750	A	X	41.8	54.3	10.3
34***.75x.75		0.750	A	X	12.5	114	15.3
35***	1X1	0.750	A	Z	253	---	---
36***	1X1	0.750	E	Z	177	---	---

A - Epoxysiloxane with an Epoxy Equivalent Weight of 865  
coated onto 0.085mm thick cast polypropylene

X - Test Tape X.

B - Epoxysiloxane with an Epoxy Equivalent Weight of 445

coated onto 0.089mm thick cast polypropylene

Y - Test Tape Y.

C - Wacker-Chemie "VP-1530" coated onto 0.085mm thick cast polypropylene film.

Z - Test Tape Z.

#5336 - Foam Tape #5336 available from 3M Company, St. Paul, MN.

#5337 - Foam Tape #5337 available from 3M Company, St. Paul, MN.

D - Wacker Dehasive™ 920 thermally cured silicone coated onto 0.085 mm cast polypropylene film.

E - Epoxysiloxane with an Epoxy Equivalent Weight of 865 coated onto 0.040 thick biaxially oriented polypropylene film.

\* - Comparative is fully coated with silicone

\*\* - Heat Aged Release Value Test is conducted as described above except that the samples were aged for 7 days at 70°C.

\*\*\* - Grid pattern in which the pattern size indicates the islands which are uncoated and the bridges form the grid which is silicone coated.



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What is Claimed is:

1. A primerless release article comprising a film substrate, having coated on at least a portion thereof, at least one layer of a printable silicone release agent, wherein said agent forms a definite geometric pattern consisting of discrete islands and bridges, said islands having dimensions of from about 0.25 mm by 0.25 mm to about 4 mm by 4 mm, said bridges forming a grid, each bridge having a width of from about 0.25 mm to about 1.0 mm.

2. A primerless release article comprising a film substrate, having coated on at least a portion thereof, at least one layer of a printable silicone release agent, wherein said agent forms a definite geometric pattern consisting of discrete islands and bridges, said islands having dimensions of from about 0.25 mm by 0.25 mm to about 3 mm by 3 mm, said bridges forming a grid, each bridge having a width of from about 0.25 mm to about 0.75 mm.

3. A primerless release article according to claim 1 wherein said islands are coated with said silicone release agent, and said bridges are uncoated.

4. A primerless release article according to claim 1 wherein said bridges are coated with said silicone release agent, and said islands are uncoated.

5. A primerless release article according to claim 4 wherein said coated islands have dimensions of from about 0.5 mm by 0.5 mm to about 3 mm by 3 mm.

6. A primerless release article according to claim 1 wherein said silicone release agent is uv-curable.

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7. A primerless release article according to claim 4 wherein said silicone release agent is a crosslinkable silicone.

5 8. A primerless release article according to claim 1 wherein said islands are in the form of squares.

10 9. A process for providing a primerless release article consisting of coating at least one surface of a substrate with at least one printable silicone release agent in a definite geometric pattern, such that discrete islands and bridges are formed, said islands being coated with said silicone release agent, and said bridges being uncoated.

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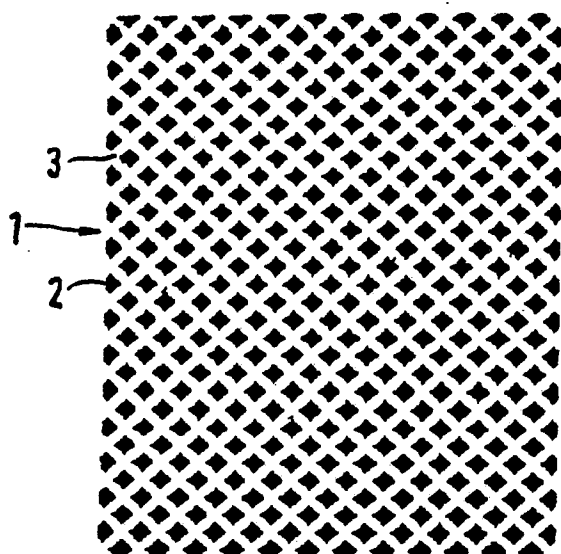


FIG. 1

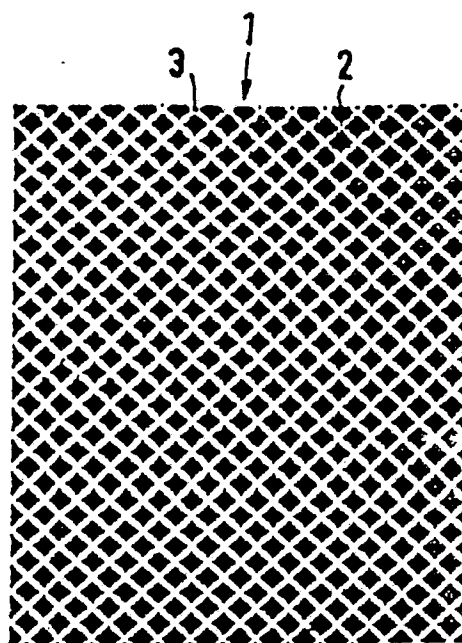


FIG. 2

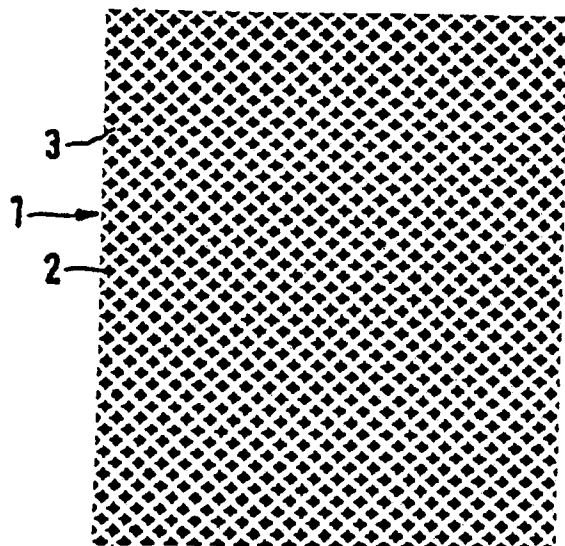


FIG. 3

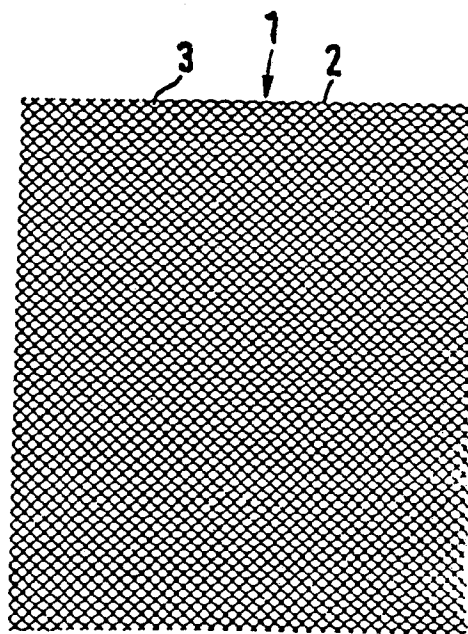


FIG. 4

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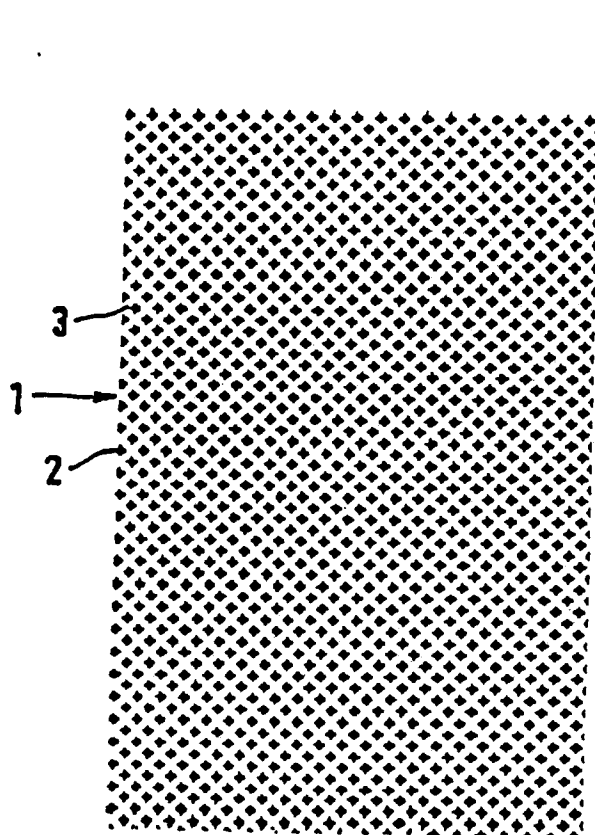


FIG. 5

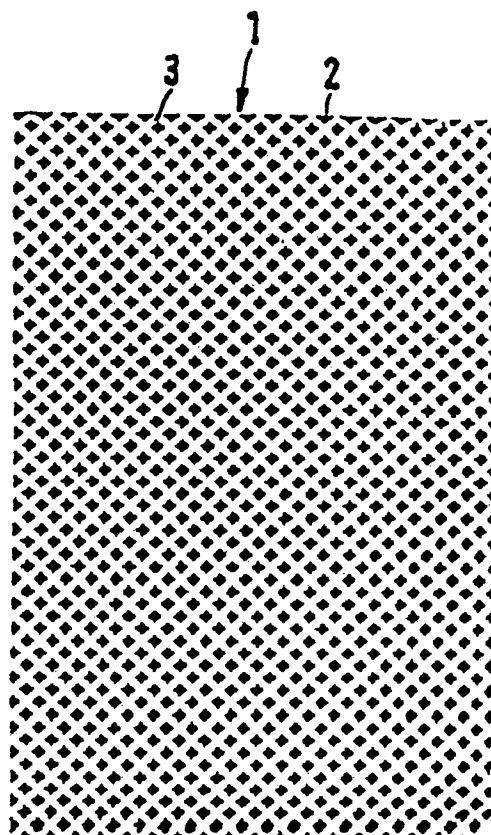


FIG. 6

# INTERNATIONAL SEARCH REPORT

International Application No. **PCT/US 89/05804**

<b>I. CLASSIFICATION OF SUBJECT MATTER</b> (if several classification symbols apply, indicate all) <sup>1</sup>		
According to International Patent Classification (IPC) or to both National Classification and IPC <b>IPC5: C 09 J 7/02, B 32 B 7/06</b>		
<b>II. FIELDS SEARCHED</b>		
Minimum Documentation Searched <sup>2</sup>		
Classification System	Classification Symbols	
<b>IPC5</b> <b>B 05 D; B 32 B; C 09 J</b>		
Documentation Searched other than Minimum Documentation to the Extent that such Documents are Included in the Fields Searched <sup>3</sup>		
<b>III. DOCUMENTS CONSIDERED TO BE RELEVANT <sup>1</sup></b>		
Category <sup>4</sup>	Citation of Document, <sup>11</sup> with indication, where appropriate, of the relevant passages <sup>12</sup>	Relevant to Claim No. <sup>13</sup>
P,A	EP, A1, 0307578 (BEIERSDORF AKTIENGESELLSCHAFT) 22 March 1989, see claim 8	1-8
P,X	--	9
A	EP, A1, 0209103 (NYFFELER, CORTI AG) 21 January 1987, see column 2, line 17 - line 41; column 3, line 12	1-8
A	US, A, 2539420 (J. E. HAZELTINE, JR., ET AL) 30 January 1951, see column 2, line 30 - line 47; column 3, line 30 - line 35	1-8
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<div style="display: flex; justify-content: space-between;"> <div style="width: 45%;"> <p><sup>4</sup> Special categories of cited documents: <sup>10</sup></p> <p>"A" document defining the general state of the art which is not considered to be of particular relevance</p> <p>"E" earlier document but published on or after the international filing date</p> <p>"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)</p> <p>"O" document referring to an oral disclosure, use, exhibition or other means</p> <p>"P" document published prior to the international filing date but later than the priority date claimed</p> </div> <div style="width: 45%;"> <p>"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention</p> <p>"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step</p> <p>"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.</p> <p>"Z" document member of the same patent family</p> </div> </div>		
<b>IV. CERTIFICATION</b>		
Date of the Actual Completion of the International Search	Date of Mailing of this International Search Report	
11th April 1990	<b>27 APR 1990</b>	
International Searching Authority	Signature of Authorized Officer	
<b>EUROPEAN PATENT OFFICE</b>	<b>Mme N. KUIPER</b>	

ANNEX TO THE INTERNATIONAL SEARCH REPORT  
ON INTERNATIONAL PATENT APPLICATION NO. PCT/US 89/05804

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This annex lists the patent family members relating to the patent documents cited in the above-mentioned international search report.  
The members are as contained in the European Patent Office EPP file on 28/02/90  
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Patent document cited in search report	Publication date	Patent family member(s)	Publication date
EP-A1- 0307578	22/03/89	DE-A- 3727078 JP-A- 1090279	23/02/89 06/04/89
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